

Microwave Absorption Properties Of Magnetic Perovskite/Epoxy Based Composites:A Predictive Approach

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Abstract:

Magnetic perovskite compounds have enhanced dielectric and magnetic properties thereby providing good microwave absorption properties with frequency tunability. Hence, in this work La based magnetic perovskite fillers are selected using theoretical model and tested further for their overall microwave absorption characteristics. The theoretical model used in the study utilizes polarization energy of individual elements as a base parameter and to predict compound formulation by applying the empirical balancing equation. If the selected elements satisfy the given equation, the predicted compound can show efficient microwave absorption. For the experimental analysis, the perovskite fillers are combined with epoxy resin to form composite samples which were then used to measure the reflection loss in a far field set up. The measured reflection loss was well above -20 dB for the selected compounds verifying the theoretical analysis. A shift towards in frequency from 8 to around 4 GHz was observed as the thickness for the sample is increased from mm to cm range. The samples have also shown high reflection loss on and around a certain frequency value suggesting a good frequency selection ability. The comprehensive study highlights the use of perovskites as an affordable and efficient alternative to carbon based microwave absorbers.

Keywords: Perovskite, Composite, Reflection Loss, Polarization energy, Balancing Equation, Resonance Frequency.

1. Introduction

Perovskite based microwave absorbers have gained immense popularity in the recent years owing to their enhanced dielectric and magnetic properties. Moreover, their ease of fabrication, cost effectiveness and easy accessibility have made them a viable commercial alternative to carbon based microwave absorbers. On comparing the carbon based microwave absorbers to perovskite based microwave absorbers, the latter provides advantages in light weighted structures as the thickness required for perovskite absorbers to reach the required microwave absorption efficiency is much less than the former. The decreased thickness can be attributed to the tunable characteristics of perovskite compounds providing both dielectric and magnetic dissipation losses across the absorber. For the stated reasons, a lot of research can be seen focusing on developing new and varied kinds of perovskite based microwave absorbers with all possible element configurations [1,2,3]. This vast domain of research data emphasizes the fact of the potential and choices that perovskite based compound can offer for the possible application requirements. However, in order to harness this flexibility that these three dimensional metallic oxide ceramic presents there has to be a theoretical and mathematical framework that can

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encompass all the properties and critical parameters involved in the absorption mechanism. This way a prediction methodology can be used to form different compound configurations as per the required application saving the experimental resources. In the current work the above approach has been used to study and determine the reflection loss of magnetic perovskite compounds. Firstly, a theoretical model is used to verify the selected compounds using the polarization energies and the balancing equation. After the theoretical verification the compounds were then fabricated as composites and tested for their reflection ability. The processes involved are each explained with detail in the coming sections. The given approach enables to understand the role and effect of the dielectric and magnetic phenomena working in the back of the absorption mechanism.

The various properties like the permittivity, permeability, polarization energy, valency factor, nucleus charge all are mathematically accounted for in the determination of the compound creating a deeper understanding of how these perovskite fillers responds to the incident frequencies. The proposed work displays a unique approach of integrating theoretical findings with the experimental results.

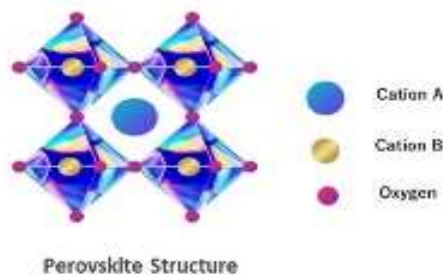


Fig. 1: Perovskite Structure

2. Modelling

The theoretical model used in the current work is derived from the previous work [4,5] where a detailed mathematical model is developed to conclude a set of balancing equation for dielectric and magnetic based perovskite compound and predict the polarization compatibility of different elements towards each other to give efficient microwave absorption. The balancing equation derived for the magnetic based compound is as given below:

$$(1) \quad \frac{P_{E_1} - P_{E_2}}{P_{E_1}} \times Ne \times Z = \sum \text{magnetic dipole moment} \times n \times \text{valency}$$

Where P_{E_1} , P_{E_2} is the polarization energy of Cation A and Cation B respectively. Ne is the number of electrons in oxygen, Z is the atomic number of oxygen, n is the atomic ratio of the magnetic element present in the compound. The given empirical equation defines the nature and response of different cations in a perovskite compound. Fig. 1 depicts a basic ABO_3 type perovskite compound comprises of two cations and an anion in which case there are two polarizing cations and an anion present in proximity to each other in an incident electromagnetic field affecting the overall the overall attenuation [6,7].

To describe it further, firstly the role of the polarization energy must be explained. The polarization power of an atom is a measure of its ability to polarize that is to separate the positive and negative charges to form an electric dipole. The formation of an electric dipole attenuates the incident frequency by forming an attenuating electric field with respect to the created dipole. In the example shown in Fig. 2, there are three electromagnetic fields

present, one is for the incident wave, the other is the cationic electric field while the third is around the polarized anion. The induced local electric field is dominated by two factors one is the polarization energy of the cations, second is their respective valency which affects the charge on the cations hence the strength of the created dipole. Now if the polarization power of the cations is equivalent to polarizing power of the anion maximum attenuation of the incident field can be obtained.

However, if there is an unbalance between the polarizing power one will dominate the other thus deforming the other local field decreasing the overall attenuation in dielectric based perovskite absorbers.

But in case of magnetic based perovskite the decrease is compensated with the attenuation presented by the additional magnetic field formed due to the presence of spin and orbital dipole moment. The physical significance of the balancing equation helps in portraying the creation and effect of the local electric and magnetic field in the attenuation of the incident electromagnetic field. From the above example, it is evident that because of the presence of both dielectric and magnetic losses, a magnetodielectric perovskite absorber can give higher reflection loss than just the dielectric c absorber.

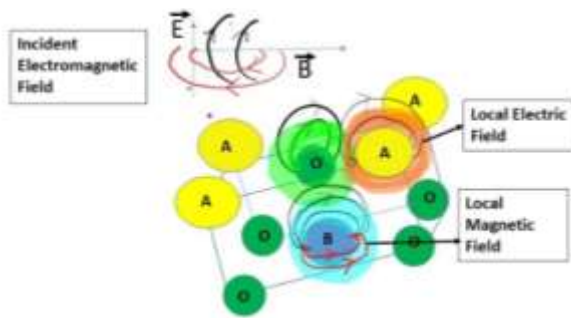


Fig. 2: Loss Mechanism of absorbed waves in microwave absorbers

2.1 Loss Mechanism

The loss mechanism in a composite microwave absorber consists of many factors. It is a cumulative effect of number of physical concepts[8]. The important ones as shown in Fig. 3, are the creation of electric and magnetic dipoles, relaxation of the dipoles, eddy current losses due to the conductivity, multiple internal reflections at charge interfaces, interfacial polarization, wave entrapment at the defect boundaries and final is the phase cancellation due to quarter wave thickness[9,10]. If all of these phenomenon are combined together a sufficient amount of reflection loss can be achieved equal to or greater than -20 dB signifying an absorption rate of around or more than 99% of the incident wave power.

2.2 Modelling

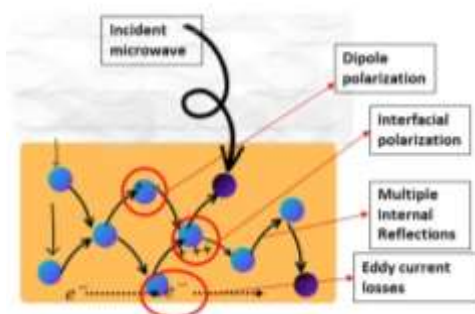


Fig. 3: Multiple Reflection and wave

For sample modelling eq. (1) is used to come up with three different perovskite fillers having comparable polarization energies and overall magnetic moment. Fig. 4 is used to show the application of the balancing equation to model the given compounds.

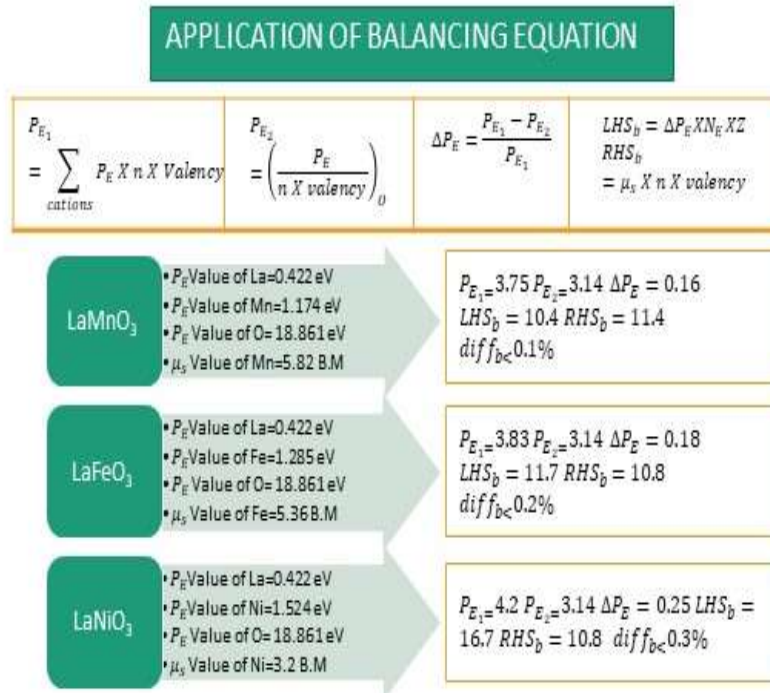


Fig. 4: Prediction of Perovskite based microwave absorbing compounds using balancing equation

3. Methods

3.1 Fabrication

The Raw materials used for the matrix system are biphenyl epoxy resin, diethyl toluene diamine (DETDA) hardener and LaMnO₃, LaFeO₃, LaNiO₃ micron sized perovskite filler powders. The raw materials are procured from Yaavik Materials & Engg. Pvt. Ltd. The three were mixed at a weight ratio of 100:24, and then manually mixed with specified weight ratio of given perovskite filler. The mixture is then poured into the die mold and cured at curing was carried at 100 deg. C for 1 hour, 135 deg. C for 3 hours with final sample as given in Fig. 5.

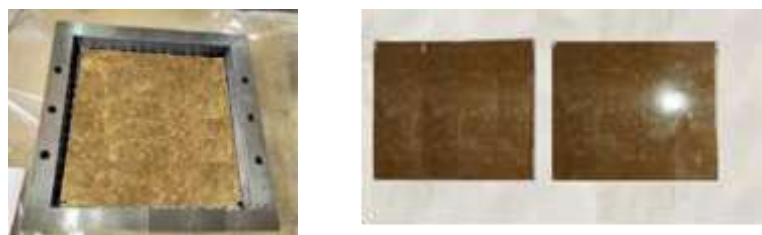


Fig. 5: Prepared Composite Samples cured and dried

The sample parameters including the filler ratio, thickness and material can be found in Table I. The thickness of sample is increased with the filler ratio. The cured samples are tested for the reflection loss at the far field measurement facility in Advanced System Laboratory, DRDO.

TABLE I : Prepared Composite Samples

Sample	Filler	Filler Ratio	Thickness (mm)	Area (mm ²)
SP1	LaMnO ₃	0.3	3	300
SP2	LaMnO ₃	0.5	3	300
SP3	LaMnO ₃	0.8	3	300
SP4	LaMnO ₃	0.5	15	300
SP5	LaFeO ₃	0.3	3	300
SP6	LaFeO ₃	0.5	3	300
SP7	LaFeO ₃	0.8	3	300
SP8	LaFeO ₃	0.5	15	300
SP9	LaNiO ₃	0.3	3	300
SP10	LaNiO ₃	0.5	3	300
SP11	LaNiO ₃	0.8	3	300
SP12	LaNiO ₃	0.5	3	300

3.2 Measurement

The sample properties are measured in a Vector Network Analyzer (VNA) (M/s Keysight, Model PNA-N5224A), using free space measurement set up. The fixtures for holding the transmitting and the receiving antennas are mounted on vibration free platform by suitable clamp fixtures. The samples is mounted at the center facing the antennas perpendicularly. The calibration is carried out by Through-Reflect-Line measurement of the antennas without sample. After carrying out the Through-Reflect-Line calibration measurement for the reflection loss are taken. Aluminium foil is used as reflective conductive plate to back the samples. The whole frequency range from 2 GHz to 13 GHz was covered by using different adapter combinations which gives a good idea of near field reflection loss (S_{11}). Out of them the average value is considered.

The measured reflection loss against the frequency range for the different samples are given below in Fig. 6(a)-6(d). The reflection loss and the resonance frequency(f_r) captured for different filler ratio and thickness is tabulated in Table II

4. Result and Discussion

The perovskite filler ratio is kept around 0.3%, 0.5% and 0.7% for a thickness of 3 mm and 0.5% for a thickness of 15 mm. The plot shows the variation of reflection loss from 0 to -40 dB against a frequency range of 2-13 GHz. A comparison analysis can be seen for the three different perovskites for different fillers and thickness ratio showing a good amount of absorption. As seen from the plots, at $x=0.5$ and $d=3\text{mm}$ LaMnO₃ has given maximum amount of reflection loss of around -30.8 dB at 7 GHz frequency. The values for the other two fillers are RL=-25.3 dB at 7.6 GHz for LaFeO₃ and RL=-14.2 dB at 9.4GHz for LaNiO₃. Similarly for $x=0.3$ and $d=3\text{mm}$ LaMnO₃ has a RL value of -19.1 dB at 7.0 GHz comparable to that of LaFeO₃ which has RL=-20.2 dB at a frequency of 7.6 GHz, and LaNiO₃ with RL=-18 dB at 9.4 GHz. Looking at the results it can said that the absorption is maximum at the mid value of filler ratio. Also Mn has shown the maximum amount of absorption owing to its high polarization energy and magnetic moment providing high permittivity and permeability to the sample. Mn is followed by Fe which have a slightly lower magnetic moment and is in turn followed by Ni which have a lower polarization energy and magnetic moment hence, a lower amount of reflection loss. Moving to $x=0.8$ and $d=3$ mm the reflection loss decreases with a value of RL=-15.6 dB at 7.0 GHz for LaMnO₃, RL=-12.4 dB at 7.6 GHz for LaFeO₃ and RL=-11 dB at 9.4 GHz. Consecutively,

the reflection loss at an increased thickness of 15 mm and $x=0.5$ is found around $RL=-25.2$ dB at 3.9 GHz for $LaMnO_3$, $RL = -26.7$ dB at 3.9 GHz for $LaFeO_3$ $RL= -19.4$ dB at 5.7 GHz for $LaNiO_3$.

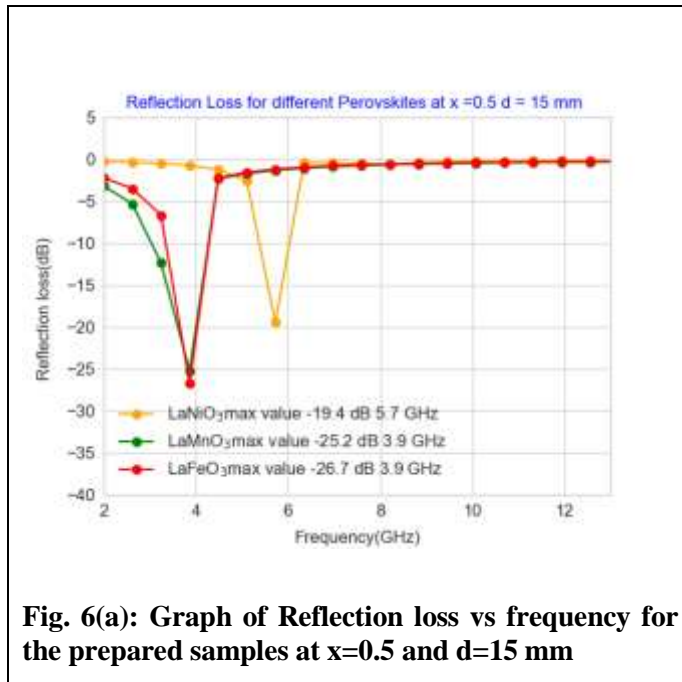


Fig. 6(a): Graph of Reflection loss vs frequency for the prepared samples at $x=0.5$ and $d=15$ mm

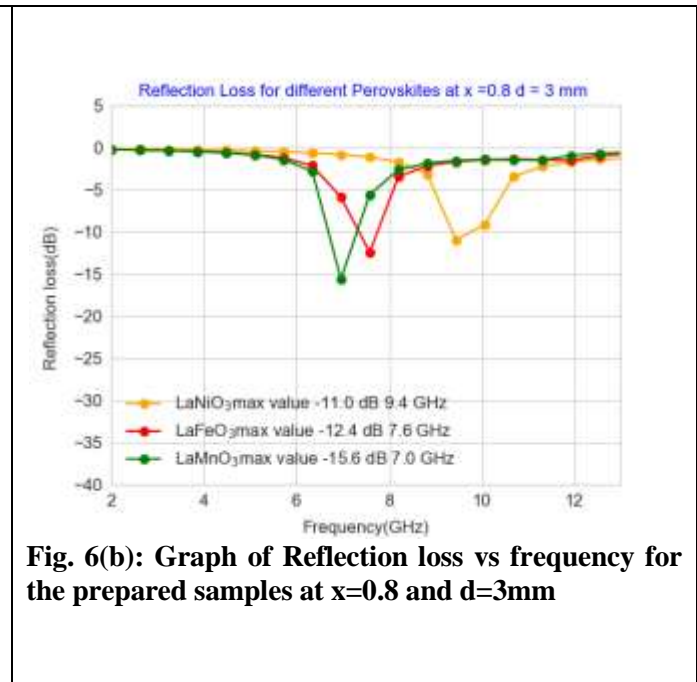


Fig. 6(b): Graph of Reflection loss vs frequency for the prepared samples at $x=0.8$ and $d=3$ mm

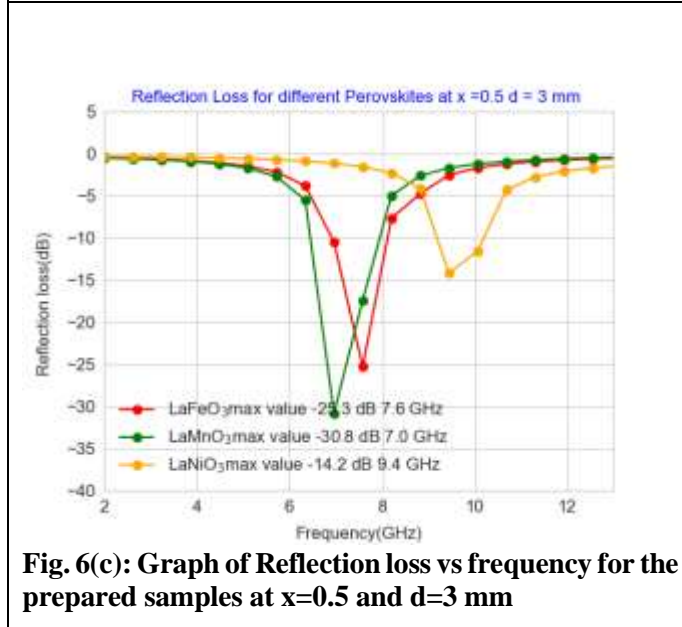


Fig. 6(c): Graph of Reflection loss vs frequency for the prepared samples at $x=0.5$ and $d=3$ mm

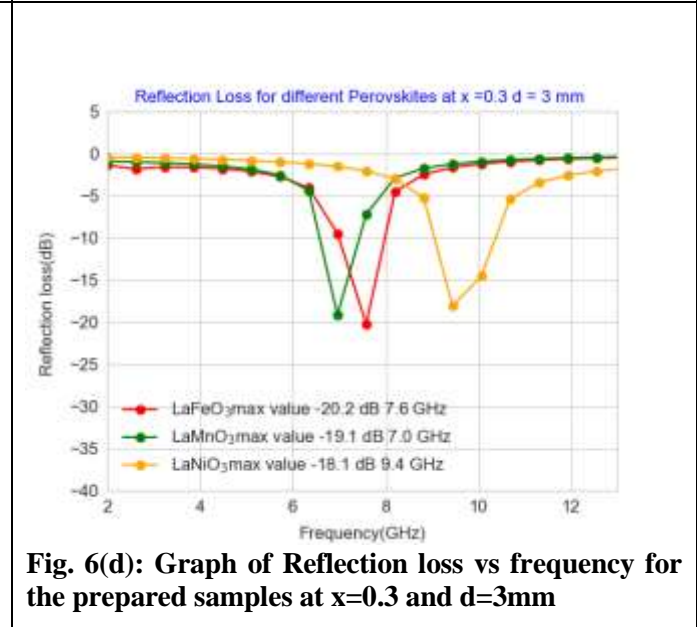


Fig. 6(d): Graph of Reflection loss vs frequency for the prepared samples at $x=0.3$ and $d=3$ mm

Table II : Reflection Loss data

Sample	RL(dB)	f_r (GHz)
SP1	-19.1	7
SP2	-30.8	7
SP3	-15.6	7
SP4	-25.2	3.9
SP5	-20.2	7.6
SP6	-25.3	7.6
SP7	-12.4	7.6
SP8	-26.7	3.9

SP9	-18.1	9.4
SP10	-14.2	9.4
SP11	-11.0	9.4
SP12	-19.4	5.7

From the data above, the microwave absorption efficiency of perovskite based absorbers can be understood. When the cationic and anionic polarization and magnetic energy balances the compound can provide good microwave absorption. From the above results, it can be seen that the higher magnetic moment values of Fe and Mn in LaFeO₃ and LaMnO₃ contributes mainly towards their higher value of reflection loss than Ni in LaNiO₃. The observation along with the given theoretical statement of, “a magnetodielectric perovskite absorber can give higher reflection loss than just the dielectric absorber” provided in previous section explaining the role of the dielectric and magnetic dissipation losses in overall microwave absorption.

5. Conclusion

The proposed work, not only validates the use of balancing equation for the prediction of magnetic perovskite based microwave absorber but also brings out their absorption response characteristics. All the predicted compounds has satisfied the balancing equation by having a comparable polarization energy on both the cationic and the anionic side. Further, these compounds have shown a fairly good amount of absorption response at their respective resonance frequencies not only proving the theoretical prediction but bringing out some other key points that can be concluded from their response variation. Looking at the said variations, it is seen that with a thickness in mm range the resonance frequency lies in the 7-8 GHz range showing a maximum amount of variation with the filler ratio of $x=0.5$. As the thickness is increased to cm range the resonance frequency shifts towards a lower range of around 2-4 GHz. The reason for this variation is the change in the quarter wave thickness for the sample. The amount of reflection loss is proportional to the polarization energy and the magnetic moment of the respective cation.

6. References

- [1] Dou, Zhanming, et al. "Understanding microwave dielectric properties of $(1-x)$ CaTiO₃- x LaAlO₃ ceramics in terms of A/B-site ionic-parameters." *Journal of Advanced Ceramics* 6 (2017): 20-26.
- [2] Jia, Zirui, et al. "Laminated microwave absorbers of A-site cation deficiency perovskite La_{0.8}FeO₃ doped at hybrid RGO carbon." *Composites Part B: Engineering* 176 (2019): 107246.
- [3] Bai, Jing, et al. "Efficient single and bi-layer absorbers of CaTiO₃ micro-cubes and polypyrrole nanotubes composites for enhanced microwave absorption in X and Ku band." *Ceramics International* 48.9 (2022): 11953-11961.
- [4] Arya, Aayushi, and G. V. V. Sharma. "Prediction of material composition for microwave absorption through mathematical modelling." *Journal of Physics Communications* 5.12 (2021): 125002.
- [5] Arya, Aayushi. "Development of a Graphical User Interface for Reflection Loss Calculation in Perovskite-RGO based Microwave Absorbing Composites." *Engineering, Technology & Applied Science Research* 13.1 (2023): 9991-9996.
- [6] Dai, Sisi, et al. "Interfacial polarizations induced by incorporating traditional perovskites into reduced graphene oxide (RGO) for strong microwave response." *Dalton Transactions* 48.7 (2019): 2359-2366.
- [7] Liu, Xiang, et al. "Enhanced microwave absorption properties by tuning cation deficiency of perovskite oxides of two-dimensional LaFeO₃/C composite in X-band." *ACS Applied Materials & Interfaces* 9.8 (2017): 7601-7610.

- [8] Panwar, Ravi, et al. "Design of ferrite–graphene-based thin broadband radar wave absorber for stealth application." *IEEE Transactions on Magnetics* 51.11 (2015): 1-4.
- [9] Singh, Sandeep Kumar, M. J. Akhtar, and Kamal K. Kar. "Impact of Al₂O₃, TiO₂, ZnO and BaTiO₃ on the microwave absorption properties of exfoliated graphite/epoxy composites at X-band frequencies." *Composites Part B: Engineering* 167 (2019): 135-146.